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Improvement of IBAD MgO Template Layers on Metallic Substrates for YBCO HTS Deposition

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***Abstract*—We present our results to improve ion beam assisted deposition (IBAD) of magnesia (MgO) templates for subsequent $YBa_2Cu_3O_{7-\delta}$ (YBCO) deposition. The substrate surface roughness has a significant effect on the initial nucleation texture of IBAD MgO films. We have found that reduction in our substrate surface roughness to $\sim$ 3.5 nm has resulted in better in-plane texture for IBAD MgO films than those deposited on rougher metal substrates. We have further improved the IBAD MgO deposition parameters by using an in situ reflected high-energy electron diffraction (RHEED) analysis tool that allows for monitoring of IBAD MgO growth. We have found a direct correlation between the RHEED generated intensity versus time curve and in-plane texture. Utilizing X-ray diffraction analysis, we have been able to determine the optimum deposition parameters to routinely grow films in batch mode that have a phi scan $\Delta\phi$ from 6–7°. Coupling the improvements in substrate preparation with optimization of IBAD MgO deposition has allowed for both batch and continuous deposition (termed c-IBAD MgO) on metallic substrates that result in superior superconducting properties. We have demonstrated that deposited meter lengths have had phi scan FWHM values from 8–9° with ±10% uniformity. Additionally, we have been able to widen the processing zone in our system and coat two, one-meter lengths simultaneously while preserving good texture quality ($\Delta\phi_{ave} \sim 8°$) and uniformity (60-80% of tape length within ±5% of $\Delta\phi_{ave}$) for both tapes.**

***Index Terms*—Coated conductor, high-temperature superconductor, ion-beam assisted deposition, magnesium oxide.**

I. INTRODUCTION

ONE OF THE drawbacks to commercialization of $YBa_2Cu_3O_{7-\delta}$ (YBCO) superconductors is the so-called "weak-link" behavior that limits polycrystalline thin film superconducting performance due to grain boundary misorientation angle [1]. This limitation is offset by the superior irreversibility in field exhibited by YBCO as compared to the Bi-based superconductors [2]. Consequently, the deposition of YBCO films on metallic polycrystalline substrates, which provide a robust platform for long-length deposition and high mechanical strength for a variety of applications, is of importance to the power industry [3].

A successful technique for depositing biaxially oriented YBCO on metallic substrates is the use of ion beam assisted deposition (IBAD) to grow a biaxially textured film of yttria-stabilized zirconia (YSZ) on mechanically polished nickel alloy tapes [4], [5]. The use of this process has resulted in the fabrication of meter-long coated conductors with critical current densities (J_c) of 1 MA/cm^2 (75 K, self field), or better, for YBCO films $\geq$ 1 μm thick[6]. The IBAD YSZ process develops texture by evolutionary grain growth and requires between 500 to 1000 nm of material to achieve reasonably good in-plane texture (phi scan full-width-at-half-maximum or $\Delta\phi$ of $\sim$ 12°). For this reason, this process has been criticized for the amount of time necessary to deposit the IBAD YSZ layer, which takes $\sim$ 20 hours per meter [7]. Recently, researchers at Fujikura have deposited a 30-meter length of IBAD tape using a YSZ variant, $Gd_2Zr_2O_7$, resulting in a YBCO J_c of 0.81 MA/cm^2[8]. Even with this improvement in deposition rate, the process is still slow for realization of coated conductor industrial-scale fabrication.

We have focused intense effort on developing a material to use as a substitute for YSZ in IBAD processing that will reduce overall processing time. Wang *et al.* found that comparable in-plane texture could be achieved using magnesium oxide (MgO) as a source material [9]. They observed that IBAD MgO required only 10 nm to develop texture comparable to that of IBAD YSZ at 1 μm thickness, which, for equivalent deposition rates, translates to a process that is $\sim$ 100 times faster than IBAD YSZ. We have deposited IBAD MgO on nickel-based superalloy substrates prepared by our typical mechanical polishing method used for our IBAD YSZ depositions. Early results produced $\Delta\phi$ values for the YBCO of 7.5° but critical current densities less than 0.5 MA/cm^2 (75 K, self-field) [10]. An in-plane texture value of this magnitude should have resulted in a much higher J_c value. We concluded that other factors were contributing to the degradation of the superconductor performance.

The majority of our work over the past year has focused on optimizing IBAD MgO for subsequent YBCO deposition on technically important polycrystalline nickel-based super-alloys. In order to achieve this performance, we have developed methods to improve upon specific parameters in our deposition technique. The two most influential parameters are IBAD MgO thickness and substrate roughness. To date, we have deposited several YBCO films ($\geq$ 1.5 mm) on small-area metal substrates with IBAD MgO template films that have achieved over 1 MA/cm^2 [11].

Here, we discuss our recent results on short sample sections of metallic tapes coated with IBAD MgO as a template layer for subsequent YBCO deposition. We also discuss further work to deposit IBAD MgO on meter-length substrates in a continuous mode and present results from these experiments.

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Improvements in both uniformity and texture on continuously processed meter-lengths will also be discussed.

II. Experimental Procedure

The substrates used in this study were nickel-based alloys (Haynes 242 or Inconel 625). For batch mode studies, small area samples (1 cm by 1 cm by 0.1 mm thick) were polished using an orbital polish that allowed for control of the surface roughness (between 0.05 and 4 nm). For continuous processing, all metal substrates were 1 cm wide, 0.1 mm thick, and formed into 113-cm loops. Before deposition, the metal substrates were mechanically polished with 1 μm diamond paste to an average surface roughness of 3.5 nm. A $\sim$ 5 nm thick amorphous layer (e.g., silicon nitride) was deposited on the substrate surface using r.f. magnetron sputtering. A subsequent layer of MgO ($\sim$ 10 nm) was deposited on the amorphous layer using IBAD. For these experiments, a dual ion-beam sputtering system was used, as is discussed in detail in a previous paper [7]. Briefly, argon ions were accelerated to 750 eV with a current density of 100 $\mu A/cm^2$ using a 22 cm $\times$ 2.5 cm Kaufman ion source. The ion source incidence angle was at 45° relative to the substrate. Concurrently, a second ion gun provided the MgO vapor flux of 0.04 nm/s from a 15-cm-diameter stoichiometric MgO target. The ion-to-atom ratio was kept constant at 0.7, which reduced the effective deposition rate by 30% to 0.028 nm/s due to resputtering. This processing rate (1/4$\times$ of our reported deposition rate) was used to increase the temporal width of the processing window and allowed for greater control of the IBAD deposition for these detailed experiments. The vapor flux and the ion fluence were monitored with a quartz crystal microbalance and a Faraday cup, respectively. All IBAD depositions were performed at room temperature. In some cases, a homoepitaxial layer of magnetron sputtered MgO (100 nm thick at 500°C) was added. This thickness allowed the MgO film texture to be quantified by standard X-ray diffraction.

IBAD film growth was monitored *in situ* using Reflected High-Energy Electron Diffraction (RHEED) by collecting a spot intensity versus time (I versus t) curve that used the reflections corresponding to the (002) and (022) planes. Images were captured using kSA400 software (k-Space Associates, Ann Arbor, Michigan). All patterns were taken at a beam energy of 25 keV.

Surface roughness was measured using contact atomic force microscopy (AFM)(Thermomicroscopes, Sunnyvale, CA., Model M5) at atmosphere. All samples were scanned using a 5 μm by 5 μm area. Roughness values were calculated using the root mean square (R_{rms}) of the scanned area.

III. Results and Discussion

A series of films was deposited at thicknesses that corresponded to particular times along the RHEED I versus t curve (each thickness is represented as a data point in Fig. 1). This was done to determine any correlation between the I versus t curve and the in-plane texture. Each of the films was overcoated with a homoepitaxial layer of MgO and then analyzed using an XRD ϕ-scan. We have shown that although there is some inherent difference between the IBAD film in-plane texture and that of an overcoated IBAD film, the trend from sample to sample is preserved [12]. The XRD $\Delta\phi$ results are plotted along with the I versus t curve at the corresponding times that their individual depositions were stopped in Fig. 1. At successive points along the curve the films' $\Delta\phi$ measured from 15° at the onset of growth to a minimum value of 8° near the spot intensity maximum and then increased to 18° at $\sim$ 15 seconds past the time at which the maximum spot intensity was observed. From the results of these experiments, it appears that the best texture is achieved at this maximum intensity and that the in-plane texture drastically degrades if the IBAD MgO film is allowed to progress past this maximum intensity value. A thickness of 10 nm has been found to correspond to this maximum intensity as quantified by AFM.

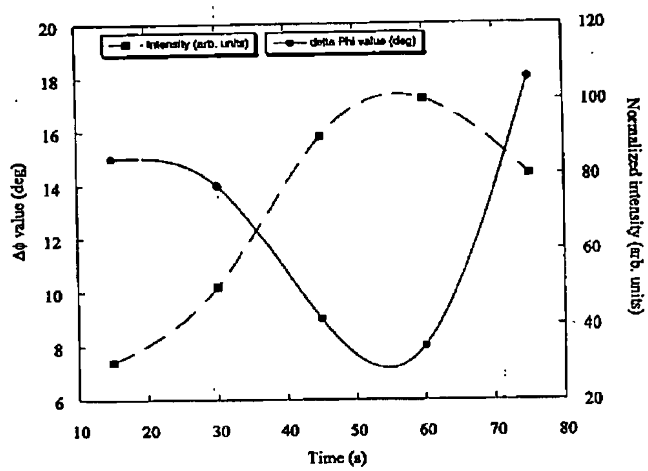

Fig. 1. A graph showing the change of in-plane texture ($\Delta\phi$) and normalized RHEED spot intensity versus time. Note that at the maximum spot intensity, a minimum in $\Delta\phi$ is observed.

If we allow the films to grow beyond the maximum in the I versus t curve, we find that some other surprising effects occur. In the present system configuration, the RHEED beam is parallel to the ion source. If, after the deposition is completed, the samples are rotated 90° with respect to the ion beam direction, the spot pattern tilts away from the ion source as the film continued to grow beyond 10 nm. The tilting progresses from $\sim$ 0° at 10 nm to $\sim$ 5° at 20 nm and to 12° at 30 nm as shown in Fig. 2. This effect was first observed by Wang and partially repeated by our group [13], [14]. As one continues to deposit IBAD MgO beyond 30 nm, the tilt continues to increase until it appears to approach an asymptotic value of $\sim$ 30° near a thickness of 100 nm. We hypothesized that as the film grows beyond 10 nm and the planes begin to tilt away from the ion beam, the ions do not channel as well along the ⟨110⟩ direction. This induces a greater amount of damage in the surrounding film thereby increasing the misorientation between grains. By stopping the growth of these films at the maximum in the I versus t curve, we have been able to deposit high-quality IBAD MgO on metallic substrates. At present, we are capable of routinely depositing films with $\Delta\phi$ between 6° and 7°[14].

Another factor that has influenced the deposition of high quality IBAD MgO films is substrate roughness. As shown in

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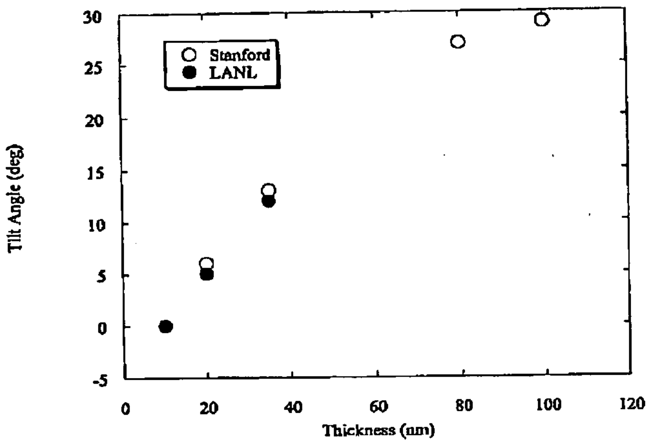

Fig. 2. Graph showing the change in MgO (200) lattice tilt as a function of IBAD MgO thickness.

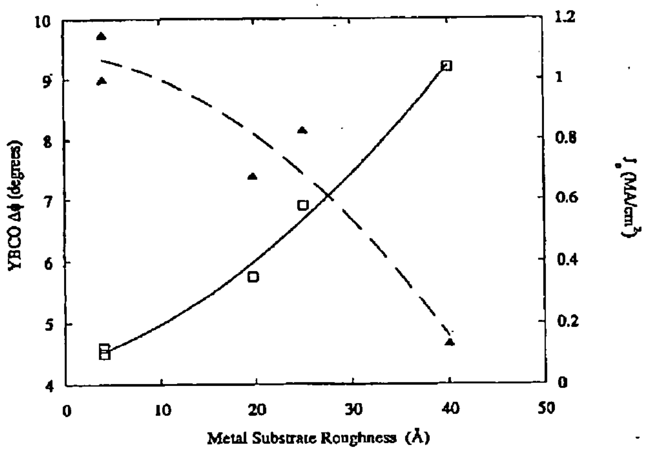

Fig. 3. The effect of metal substrate roughness on final YBCO in-plane texture ($\Delta\phi$) and superconducting critical current (J_c). A direct correlation is observed between reduction in the substrate surface roughness and both the texture and performance of YBCO on IBAD MgO templates.

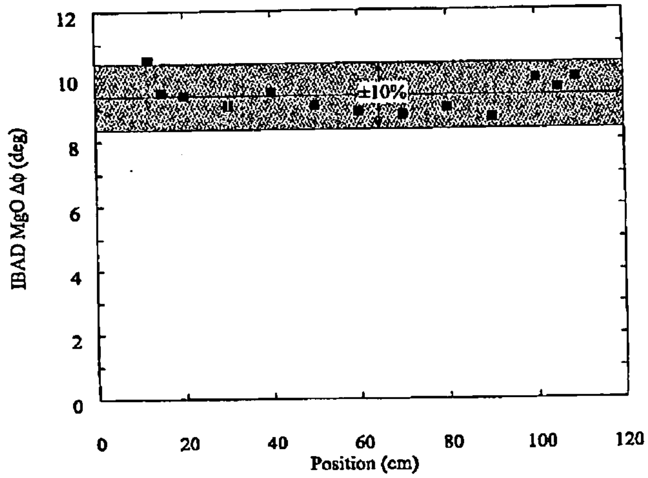

Fig. 4. A graph showing the texture uniformity of a continuously processed IBAD meter as a function of position along the tape length. The dark band indicates values of texture that are within $\pm 10\%$ of the average $\Delta\phi$ for the tape that is 9.4°.

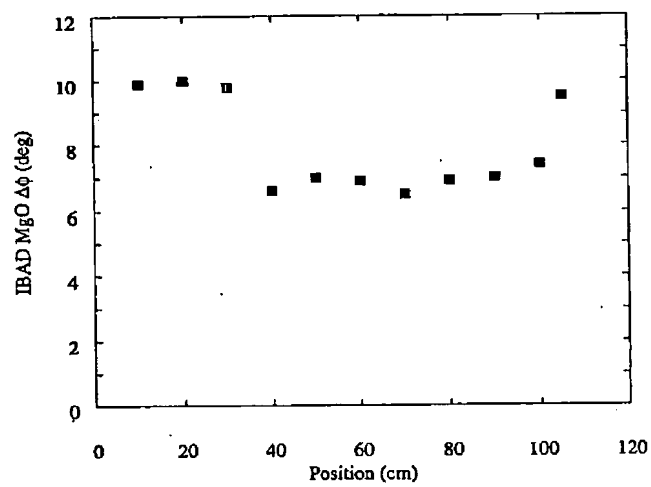

Fig. 5. In-plane texture values as a function of position for one of two simultaneously coated IBAD MgO loops. Note that over 60 cm of material has a $\Delta\phi_{ave}$ of ~ 7°.

Fig. 3, there is a direct correlation between reduction in metal substrate surface roughness and final YBCO in-plane texture as well as the corresponding critical current density (J_c). The data points shown in Fig. 3 are compiled for a recent set of experiments on batch mode samples. The substrates were prepared at different surface roughness values using the small area polishing technique described in the experimental section. All the samples used in this study had the following film architecture: YBCO/CeO_2/YSZ/homoepi-MgO/IBADMgO/a-Si_3N_4/C276. The curve fits accompanying the data points show a distinct trend that has been observed for several continuously coated IBAD MgO samples as well. As a consequence of this study, we have developed a proprietary process for preparing substrates for continuous substrates that can finish surfaces to ~ 2 nm R_{rms}.

As an example of the improvements made considering the two important factors discussed above, the superconducting transport properties were measured for a number of > 1-μm-thick YBCO deposited on continuously deposited (c-IBAD) MgO templates. In 2001 our best meter of c-IBAD MgO was fabricated with in-plane $\Delta\phi$ values along the length of the tape averaging ~ 11° [15]. The best 25-cm section (average $\Delta\phi$ 9°) from this tape was cut into 1 cm pieces and coated with YBCO using PLD. The samples were then patterned into microbridges and characterized for their superconducting transport properties using the 1 μV/cm criterion – the results of which were summarized recently [11]. The average J_c for these 24 samples was 0.89 MA/cm^2 (75 K, SF) with an average YBCO thickness of 1.44 μm. With this type of performance, a projected average I_c of 125 A/cm width was expected.

Over this past year, we have continued to refine the deposition process and improve substrate preparation to optimize the c-IBAD MgO. To this end, several meter length loops have been processed with improved in-plane texture values and uniformity. Shown in Fig. 4 is a typical meter of c-IBAD. The majority of

the meter length IBAD MgO $\Delta\phi$ values fall well within $\pm 10\%$ of the average $\Delta\phi = 9.4°$. Not only is this improvement in uniformity substantial, but the texture value has dropped by another 2° on average.

We have continued to improve our deposition technique by increasing the processing window of the c-IBAD MgO through further optimization using the RHEED I versus t curve and with enhancements in substrate surface preparation. This has resulted in the meter shown in Fig. 5. In contrast to Fig. 4, this meter (even with the scatter at the ends) has an average $\Delta\phi$ of 8°. Over 60 cm of the tape has an average $\Delta\phi$ value of 7° with the majority of the length $\Delta\phi$ within $\pm 5\%$ of this average value. Furthermore, this tape was processed alongside a second meter loop deposited upon simultaneously. The average $\Delta\phi$ for this meter was slightly higher at 8.3°. Both tapes produced well-aligned c-IBAD MgO templates.

IV. Summary and Conclusion

Two fundamental parameters have been found to greatly influence the texture values of IBAD MgO films. There exists a critical IBAD MgO thickness ($\sim$ 10 nm) and necessary substrate surface finish ($\leq$ 3.5 nm R_{rms}) that is critical to depositing high quality IBAD MgO template layers. *In situ* monitoring with RHEED I versus t curves has allowed for the expansion of the IBAD MgO processing window by determining the appropriate thickness necessary to develop highly aligned template films. We have demonstrated that it is possible to rountinely polish these Ni-alloy loops to R_{rms} values better that 3.5 nm. This ultimately results in a final YBCO film with higher performance than on previously coated IBAD MgO templates. We have demonstrated that high-quality IBAD MgO can be deposited continuously on technically relevant metallic tape. Improvements in substrate polishing, coupled with in situ RHEED, have resulted in the optimization of processing parameters and in the deposition of highly textured ($\Delta\phi_{\mathrm{ave}}$ 8–9°) c-IBAD films. These c-IBAD films have been coated with YBCO and shown to perform well. These c-IBAD MgO samples subsequently deposited in batch mode with > 1 μm thick PLD YBCO have achieved $J_c > 1$ MA/cm^2 (75 K, SF). As a further demonstration of the viability of c-IBAD MgO, two 113-cm loops have been coated with MgO resulting in well aligned material. These results confirm that c-IBAD MgO is a feasible alternative to IBAD YSZ that can have direct benefits in industrial processing of second-generation coated conductors.

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